

EOS7RN—A NEW TOUGH2 MODULE FOR SIMULATING RADON EMANATION AND TRANSPORT IN THE SUBSURFACE

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ABSTRACT

A new fluid property module, EOSRn, was developed for TOUGH2 to simulate the transport of the radon gas (^{222}Rn) in saturated-unsaturated soils. It is an enhanced version of the EOS7R module for radionuclide transport, with a source term added in the transport equation to model radon generation by emanation from radioactive decay of the soil radium (^{226}Ra) content. We implemented physical properties of this gas component in two-phase (liquid-gas) porous media, as a function of soil moisture and/or soil temperature, such as those of the diffusion coefficient, the emanation factor, the adsorption coefficient and Henry's law coefficient. To ensure that temporal and spatial numerical discretization of this nonlinear source term is effective and has been well implemented in TOUGH2, we performed comparative studies between EOS7Rn and an exact analytical solution at steady-state isothermal unsaturated conditions for many numerical experiments of one-dimensional radon transport in homogeneous and layered soil columns.

We found that the radon activity concentration profiles and flux densities calculated by EOS7Rn were in good agreement with the analytical solution for all the studied numerical experiments. Consequently, relative errors for calculated TOUGH2's radon mass balance and flux densities were very negligible. Like most other sister modules, EOSRn can simulate nonisothermal multiphase flow and fully coupled three-dimensional transport in fractured porous media. It will help in predicting the radon exhalation from highly radium-contaminated

soils and underground cavities to outdoor and indoor environments.

INTRODUCTION

Radon (^{222}Rn), a naturally occurring radioactive gas, with a half-life of 3.8 days, is produced by the radioactive decay of radium ^{226}Ra , which is in turn a decay product of the radioactive decay series of uranium ^{238}U . It occurs naturally in small concentrations in all soils, but in higher concentrations in granite, shale, and phosphates, which have higher concentrations of uranium. Being a rare gas, it usually migrates freely through faults and fractured soils, and may sink into groundwater or caves, or rise into the atmosphere.

Radon accounts for the largest proportion of our annual average radiation dose. Its greatest impact occurs when it becomes trapped and breaks down inside of buildings or caves. In caves or aerated mines, or ill-aerated houses, radon activity concentration can achieve many kilo-becquerels per m^3 of air, but can be much higher in mining contexts ($1 \text{ MBq} \cdot \text{m}^{-3}$). Studies concerning occupationally radon-exposed miners and direct observation of individuals exposed to radon in their homes provide firm scientific evidence that radon is a major environmental carcinogen. Radon exposure increases the risk of lung cancer—it is linked to approximately 5 to 12% of lung cancer deaths in France every year, according to the French Institute for Public Health Surveillance.

The important mechanisms that affect multiphase radon transport in the subsurface are advection, diffusion, radioactive decay, dissolu-

tion or phase partitioning, adsorption on soil solid grains, and emanation from materials with radium sources (Rogers and Nielson, 1991a,b). The release mechanism of radon from mineral grain to pore space is called emanation. This mechanism is described by the emanation coefficient, E , dimensionless, which is the ratio of the radon produced in the grain of the material to the radon in the pore space of that material. Its value varies between 0 and 1, and basically depends on soil grain size-distribution, porosity, and water content (Nielson et al., 1982; Sasaki et al., 2004). It is also temperature dependent.

Emanation and transport mechanisms depend to a large extent on soil moisture content. Thus, radon flux exhalation at the soil surface is strongly affected by weather conditions (Ferry et al., 2002). The adsorption effect significantly reduces radon migration fluxes in dry soils, since vapor adsorption in these cases takes place directly on the grain surfaces (Schery et al., 1989). Radon dissolution depends on temperature (Clever, 1979) and salinity (brine) as well.

Both radon diffusion and dissolution can be strongly temperature and pressure dependent in the context of geothermal reservoirs (Shan and Pruess, 2004). Modeling of radon transport in the subsurface, involving more complex phenomena occurring near the land surface or in deeper unsaturated zones, requires accurate description of hydrogeological features, physical processes, and thermodynamic properties. While few numerical simulators can study radon transport—in the context of nonisothermal flows of multiphase, multicomponent fluids in permeable (fractured and porous) media, by accounting for such mechanisms—the TOUGH numerical simulators, based on their underlying conceptualizations and methodologies, are well suited for the solution of like vadose-zone flow and transport problems (Finsterle et al., 2008).

The fluid property module, EOS7R (Oldenburg and Pruess, 1995; Pruess et al., 1999), is one among many TOUGH2 modules that can simulate two-phase, five-component flow and transport problems. The two phases are water and gas, and the five components are water, brine, air, and two radionuclides. Unger et al. (2004) studied the transport of radon into a

tunnel at Yucca Mountain using TOUGH2/EOS7R. Their study considered radon transport in a fractured tuff, but only as a radioactive tracer without a source term of radon from the tuff. Their study was limited to estimating the large-scale hydraulic properties of the fractured tuff and its implications for the operation of the ventilation system of the tunnel.

In the context of soil/rock with radium sources, the TOUGH2/EOS7R module can be suitable for modeling radon transport in the subsurface. However, the module must be enhanced to account for the soil radon emanation process and all radon-related physical properties with their dependency on soil hydrodynamic and thermodynamic properties (water saturation, salinity, temperature and pressure). For this purpose, a new TOUGH2 module, EOS7Rn, has been developed. As an alternative to the physically based Millington and Quirk (1961) (MQ) formulation for binary diffusion of gas components in each phase of an unsaturated soil, which is already implemented in TOUGH2, the empirical formulation of Rogers and Nielson (1991a) (RN), developed specifically for radon gas diffusion, has been implemented in EOS7Rn.

In addition, four new subroutines have been implemented to account for the physical properties of radon: RnHEN for calculating the water temperature and/or salinity dependent Henry's law coefficient, RnDIF0 for calculating temperature and/or pressure-dependent diffusivities in free gas and liquid phases, RnEMAN for calculating water saturation and/or temperature-dependent emanation coefficient, and RnADS for calculating water saturation and/or temperature-dependent adsorption coefficient between gas and solid phase. Within each of these four subroutines there is the possibility of choosing between different functions or adding user-specified ones if needed.

In this study, we verify the numerical discretization of the nonlinear source term of radon emanation in the transport equation and its implementation in TOUGH2/EOS7Rn. We will focus on comparative studies between this module and an exact one-dimensional analytical solution for steady-state radon transport in homogeneous and layered unsaturated soil

columns. (Note that the temperature effect on radon transport in the subsurface will be addressed in future work.)

MODULE DESCRIPTION

First, we assume that the two radionuclides are ^{222}Rn and ^{218}Po , with the latter a stable tracer component with infinite half-life value. For each gridblock element i with a given volume V_i in the flow domain, the general space-discretized form of the mass conserving equations of each component κ , using the Integral Finite Difference Method (IFDM), is given by (Oldenburg and Pruess, 1995):

$$\frac{dM_i^{(\kappa)}}{dt} = \frac{1}{V_i} \sum_j A_{ij} F_{ij}^{(\kappa)} + q_i^{(\kappa)} \quad \kappa = 1, \dots, NK \quad (1)$$

where $NK = 5$, which is the number of components; j labels the gridblocks connected to i through the surface area A_{ij} (m^2); t is time (s); $q^{(\kappa)}$ is a sink/source term ($\text{kg.m}^{-3}.\text{s}^{-1}$); $M^{(\kappa)}$ is the mass accumulation term (kg.m^{-3}); and $F^{(\kappa)}$ is the total flux term ($\text{kg.m}^{-2}.\text{s}^{-1}$). The latter results from advection and diffusion and is given, according to Darcy's and Fick's laws, by:

$$\mathbf{F}^{(\kappa)} = \sum_{\beta=g,l} \left(X_{\beta}^{(\kappa)} \rho_{\beta} \mathbf{v}_{\beta} - \phi S_{\beta} \rho_{\beta} \tau_0 \tau_{\beta} d_{\beta}^{(\kappa)} \nabla X_{\beta}^{(\kappa)} \right) \quad (2)$$

where ϕ is porosity; $\tau_0 \tau_{\beta}$ is the tortuosity that includes a porous medium dependent factor τ_0 and a coefficient τ_{β} that depends on saturation of phase β (gas and liquid), S_{β} ; ρ_{β} is density (kg.m^{-3}); $d_{\beta}^{(\kappa)}$ is the diffusion coefficient of component κ in free fluid phase β ($\text{m}^2.\text{s}^{-1}$); \mathbf{v}_{β} is the Darcy flow velocity or flux density of the fluid phase β (m.s^{-1}); and $X_{\beta}^{(\kappa)}$ is the mass fraction of component κ in phase β (-). For radon component ($\kappa = 3 = \text{Rn}$), the terms $M^{(\kappa)}$ and $q^{(\kappa)}$ are given by:

$$M^{(\kappa)} = \phi \sum_{\beta=g,l} S_{\beta} \rho_{\beta} X_{\beta}^{(\kappa)} + \rho_d \rho_g K d_g^{(\kappa)} X_g^{(\kappa)} \quad (3)$$

$$q^{(\kappa)} = -\lambda_{\kappa} M^{(\kappa)} + u_{\kappa} E(S_l) \quad (4)$$

$$u_{\kappa} = C_s^{^{226}\text{Ra}} \rho_d \left(\frac{M_{\text{Rn}}}{N_{\text{Av}}} \right) \times 10^{-3} \quad (5)$$

where ρ_d is the rock dry bulk density (kg.m^{-3}), equal to $(1 - \phi)\rho_R$, with ρ_R is the rock solid grain density (kg.m^{-3}); λ_{κ} is the radioactive decay constant of radon ($2.1 \times 10^{-6} \text{ s}^{-1}$); $C_s^{^{226}\text{Ra}}$ is the rock radium activity mass content (Bq.kg^{-1}); M_{Rn} is the molecular weight of radon (222 g.mol^{-1}); N_{Av} is Avogadro's number ($6.022 \times 10^{23} \text{ mol}^{-1}$); and E is the emanation coefficient (-). In (3), we use the gaseous phase (g) as a reference to describe the adsorption isotherm for radon, unlike in the Oldenburg and Pruess (1995) approach. Hence, radon gas-component partitions only between the gas phase and soil solid grains. As in TOUGH2, we assume a partition phase of radon according to Henry's law:

$$P_{\text{Rn}} = K_H x_l^{\text{Rn}} \quad (6)$$

where P_{Rn} is the partial pressure of Rn (Pa); x_l^{Rn} is the mole fraction of the dissolved Rn (-); and K_H is the Henry's law coefficient (Pa).

We use the same approach of Oldenburg and Pruess (1995) for time discretization of (1), by using two variable time-weighting parameters for decay and emanation terms. This helps to alternate between Crank-Nicolson and a fully implicit scheme for both terms.

Emanation Coefficient

RnEMAN calculates the radon emanation coefficient E in (4) as a function of the soil water saturation S_l , using the widely used linear model of Nielson et al. (1982):

$$E = \begin{cases} E_w S_l / S^* + E_a (1 - S_l / S^*) & \text{if } S_l \leq S^* \\ E_w & \text{elsewhere} \end{cases} \quad (7)$$

where E_w and E_a are the emanation coefficients at saturation and at dryness (-), and S^* is the minimum water saturation on the plateau of an emanation–water saturation curve. Equation (7) has been shown to be very accurate for describing radon-emanation laboratory data in soils with different soil textures.

Adsorption Coefficient

RnADS calculates the adsorption or distribution coefficient $K d_g$ in (3) according to the empirical relationship of Rogers and Nielson (1991b):

$$K d_g = K d_g^0 e^{-b S_l} \quad (8)$$

where Kd_g^0 is the distribution coefficient at dryness ($\text{m}^3 \cdot \text{kg}^{-1}$) and b is a correlation constant, generally lying between 10 and 15 but can be significantly higher. Equation (8) assumes that Kd_g generally decreases rapidly with increasing water saturation and that adsorption on wet surfaces is negligible. Rogers and Nielson (1991b) showed that (8) can best fit the Kd_g data of Schery et al. (1989) measured at the ambient temperature for different soil textures.

Henry's Law Coefficient

The temperature-dependent Henry's law coefficient of radon is computed by RnHEN using the Crovetto et al. (1982) (CRO82) model:

$$K_H = 10^9 \cdot e^{a_0 + a_1/x + a_2/x^2 + a_3/x^3} \quad (9)$$

where $x = 10^{-3}T$, with T is temperature (K). No K_H -data for radon are available above 100°C . However, the four regression coefficients in (9) (Table 1) were obtained by fitting the Clever (1979) data for Rn to (9), for temperatures below 100°C (Figure 1).

Table 1. Best fit regression coefficients to CRO82 model for Rn K_H Clever79-data

	a_0	a_1	a_2	a_3
Rn	-23.5229	20.5167	-5.32751	0.374797

Figure 1 shows a comparison between CRO82 and the empirical model of Yaws et al. (1999) (Yaws99). Both models give satisfactory results for noble gases for temperatures below 100°C . As expected, the decrease in Henry's law coefficient with the increased molecular weight of noble gases is fulfilled, regardless of the model used.

Diffusion Coefficient in Soil Fluid Phase

Tortuosity factor

The unsaturated soil tortuosity factor ($\tau_0\tau_\beta$ in (2)), is modeled by the well-known formulation of Rogers and Nielson (1991a):

$$\tau_0\tau_g = \phi e^{-6\phi S_l - 6(S_l)^{14}\phi} \quad (10)$$

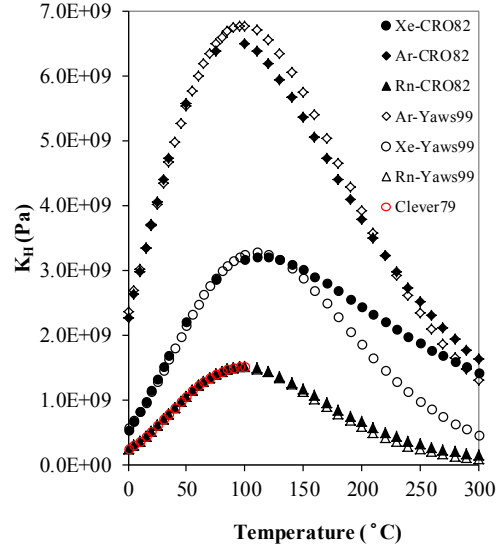


Figure 1. Best fit for Clever (1979) radon data to the CRO82 model and comparison with Yaws99 model.

where τ_0 is assumed equal to the soil porosity. Radon diffusion in soil liquid is assumed negligible in that case. Equation (10) is an empirical case obtained through the best fit to $\tau_0\tau_g d_g^{(Rn)}$ for 1073 experimental data points of diffusion coefficients measured on natural soils. As reported by Rogers and Nielson (1991a), MQ-formulation cannot be suitable for radon diffusion through earthen materials, since it considers diffusion through the pore air only, and ignores pore air-water interactions. Figure 2 shows the diffusion coefficient of radon as a function of water saturation at ambient temperature for soil with porosity $\phi = 0.4$, using both formulations, MQ and RN. The MQ-formulation is more diffusive than the RN-one with decreasing soil water saturation.

Gas phase

We use the same theoretical approach applied by Shan and Pruess (2004) for noble gases to calculate the radon diffusion coefficient in pure air and saturated water vapor ($d_\beta^{(\kappa)}$ in (2)). This approach is based on the Chapman-Enskog theory of diffusion with Lennard-Jones (LJ) intermolecular potential for binary gas mixture. Table 2 shows values of molecular weights M_{AB} and values of LJ-parameters ϵ_{AB} and σ_{AB} used for binary gas mixtures Rn-air (i.e., A=Rn, B=air) and Rn-H₂O, with H₂O being the

saturated water vapor. The latter have been calculated using values $\varepsilon_{\text{Rn}} = 283 \text{ K}$ and $\sigma_{\text{Rn}} = 4.36 \text{ \AA}$ for Rn, as taken from Gopal (1962). Also shown in Table 2 are parameter values for diffusion of air and two nobles gases (Ar, Xe) in H_2O (Shan and Pruess, 2004). The LJ-parameters of Rn- H_2O mixture are in reasonable agreement with the findings of these authors. Indeed, the values of ε_{AB} and σ_{AB} increase with increasing noble gas weight (Table 2), and inversely for radon diffusion in H_2O (Figure 3).

For groundwater flow and transport problems, temperature generally fluctuates around the ambient temperature, with air the dominant component in gas phase. Thus, it is advised to use LJ-parameters for the Rn-air mixture (Table 2).

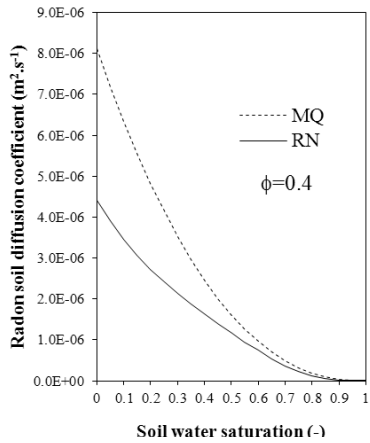


Figure 2. Radon diffusion coefficient in an unsaturated soil with porosity $\phi = 0.4$, using MQ and RN formulations.

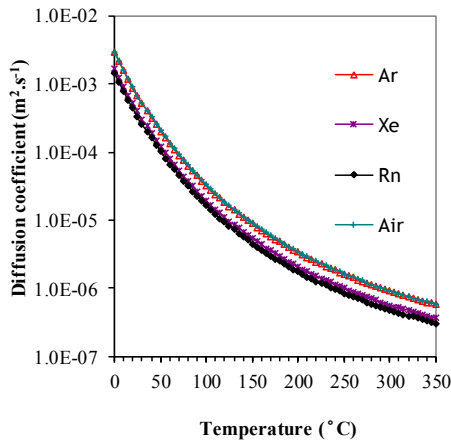


Figure 3. Diffusivity of air, Ar, Xe, and Rn in saturated water vapor.

Table 2. LJ-parameters for binary gas mixtures.

A-B	M_{AB}	σ_{AB}	$\varepsilon_{\text{AB}}/k$
Ar- H_2O	0.28859	3.0915	274.7527
Xe- H_2O	0.251242	3.344	432.3218
Rn- H_2O	0.244971	3.5005	478.5136
Rn-Air	0.197556	4.0355	149.1436
Air- H_2O	0.30005	3.176	252.181

Liquid phase

Two mathematical formulations are proposed for estimating the radon diffusion in free water as a function of temperature. The first alternative is the default empirical model of Hayduk and Minhas (1982) (HM82); the second alternative is the Wilk and Chang (1955) (WC55) theoretical model. Figure 4 shows Broecker and Peng (1974) measured data of Rn diffusivities in free water, against the two proposed models. Both models overestimate measured data with increasing temperature. The HM82 model is more accurate than the WC55, but discrepancies between them are very small compared to measurements uncertainties.

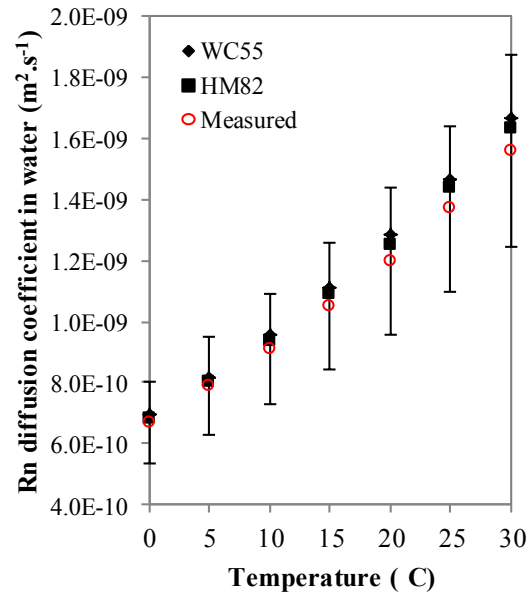


Figure 4. Comparison between WC55 and HM82 models and measured data of Broecker and Peng (1974).

MODULE VERIFICATION

Numerical Experiments

Results from the TOUGH2's numerical solution are compared with the analytical solution of Guérin (1991) (GU91) developed for one-dimensional steady-state transport in layered soil columns, with Dirichlet boundary conditions. Isothermal conditions were assumed at 25°C. $K_H = 6.06844 \times 10^8$ Pa, from (11), and the diffusion coefficient in free air and water are taken to 1.1×10^{-5} and $1.4 \times 10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$, respectively.

The column is 10 m in height with the three soil configurations generally observed in a landfill of uranium mill tailings (UMT) (1) homogeneous UMT soil, (2) two-layered soil in the sequence of a cover material (CM) over UMT (CM/UMT), and (3) five-layered soil in the sequence CM/UMT/CM/UMT/CM (CM/UMT_5L). The studied CM and UMT soils are those from the Lavaugrasse (France) landfill, which have been characterized as loamy sand and sandy silt soils, respectively (Ferry et al., 2002). Table 3 summarizes parameter values for the numerical experiments carried out on these three soil column configurations to study radon transport in single-phase gas and unsaturated conditions.

The gas pressure difference between column boundaries (Δp_g) is chosen so as to produce an upward advective transport. The steady state v_g , used as an input parameter in GU91 solution, can directly be calculated from Darcy's law for the UMT# experiments. However, for the layered case, its value is obtained from TOUGH2's numerical solution of the two-phase flow problem. For all these experiments, calculations were performed with MQ and RN-formulations. The soil profile has been assumed initially free of radon and zero, or constant radon concentration was imposed at the column boundaries ($z = 0, 10 \text{ m}$).

Table 3. Parameter values for numerical experiments of the three soil configurations.

Soil	Experiment #	S_g (-)		Δz (m)	Δp_g (Pa)
		CM	UMT		
UMT (H-UMT=10 m) and CM/UMT (H-CM=4 m, H-UMT=6 m) and CM/UMT_5L (H-CM=2 m, H-UMT=2 m)	#1	1	1	0.1	0
	#2			0.1	1000
	#3	0.5	0.5	0.02, 0.1, 0.1*	0
	#4			0.02	1000
	#5	0.75	0.25	0.02, 0.1, 0.1	0
	#6			0.02	300 ^s
	#7			0.005, 0.02, 0.01	1000
	#8	0.25	0.75	0.02, 0.1, 0.1	0
	#9			0.02, 0.01, 0.01	300 ^s
	#10			0.02, 0.01, 0.01	1000

*Different values Δz for the three soil column configurations

^sNot used for the UMT soil column

The Crank-Nicholson time discretization scheme has been used for both decay and emanation terms. The TOUGH2 numerical solution corresponds to the one achieved at steady state after solving the transient two-phase flow and transport problems. Numerical calculations are performed with regular grid spacing: $\Delta z = 0.1, 0.02, 0.01, 0.005 \text{ m}$, so as to have a Peclet number less than 2, avoiding numerical dispersion. Two values of the upstream weighting factor (WUP) for mobilities at interfaces are chosen: 0.5, 1.0. The maximum time step is 1/100 of the half-life time of radon.

Comparison between TOUGH2/EOS7Rn and GU91 analytical solution

Transport in homogeneous soil columns

Figure 5 shows a simulation example with comparison between numerical (TOUGH2: T2) and analytical (GU91) radon concentration profiles in the soil gas of an unsaturated UMT-soil column ($S_g = 0.75$), without radon adsorption. Good agreement is found between the two solutions. We obtain the same radon-concentration-profile shapes using both RN and MQ formulations, but with radon concentration higher for RN than MQ, which is in accordance with Figure 2.

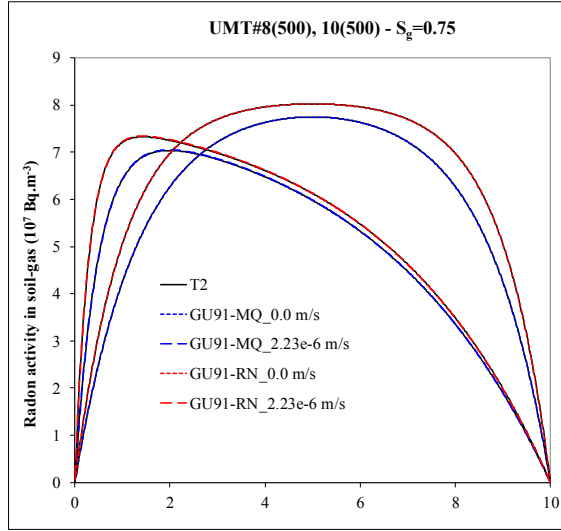


Figure 5. Numerical and analytical profiles of radon activity in the soil-gas without adsorption. Experiments UMT#8, 10 (MQ and RN).

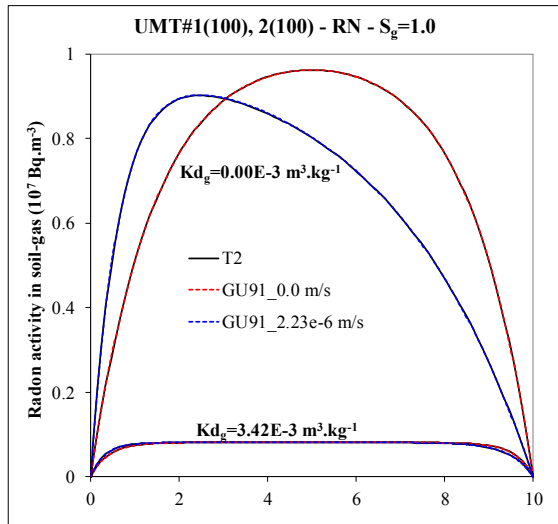


Figure 6. Numerical and analytical profiles of radon activity concentration in the soil-gas with and without adsorption. UMT#1-2 (RN).

By introducing the adsorption effect, concentration profiles are highly reduced, especially for single-phase gas conditions (Figure 6, UMT#1-2). The maximum percentage reduction of the total radon flux exhalation is 77.6% (UMT#2). For gas saturation less than 0.5, the reduction is unimportant. The radon flux at column boundaries, calculated by TOUGH2, can best fit the 1:1 curve (Figure 7), predicting that calculated by GU91. Relative errors in TOUGH2's flux density and mass balance were less than 0.8%.

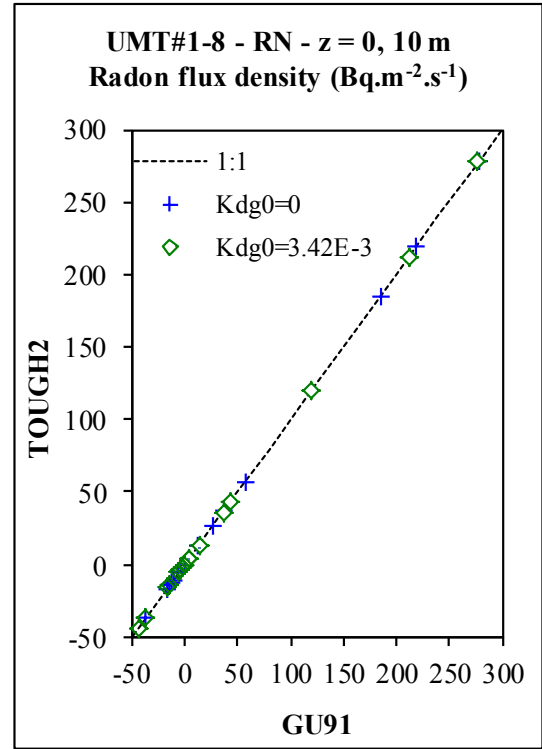


Figure 7. Numerical (TOUGH2) and analytical (GU91) radon flux densities (UMT#1-5, 7-8, 10).

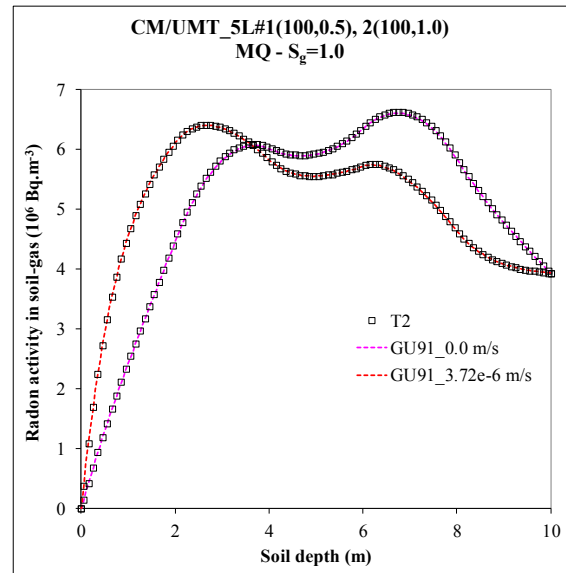


Figure 8. Numerical (T2) and analytical (GU91) profiles of radon activity in the soil-gas (CM/UMT_5L#1-2).

Transport in layered soil columns

Figures 8 and 9 show two simulation examples of radon concentration profiles calculated by TOUGH2 and GU91 for single-phase gas and unsaturated experiments CM/UMT_5L#1-2 and CM/UMT_5L#5-7, respectively. There is good agreement between the two solutions, whatever the number of soil column layers. For all experiments, the radon flux calculated by TOUGH2 at different depths is well predicted by the GU91 solution (e.g., Figure 10), regardless of the WUP-value used. However, because of the non-uniform TOUGH2's gas velocity profile, the relative error in flux density was higher for small diffusive fluxes at $z=10$ m. It exceeds 10% for CM/UMT_5L#4,9 with WUP = 1.0, but was less than 5% for WUP = 0.5. Mass-balance relative errors were less than 3.0% regardless of the WUP-value. Layering did not greatly increase the errors compared to those of the UMT# experiments, showing the mass conservation of the TOUGH2's numerical scheme IFDM.

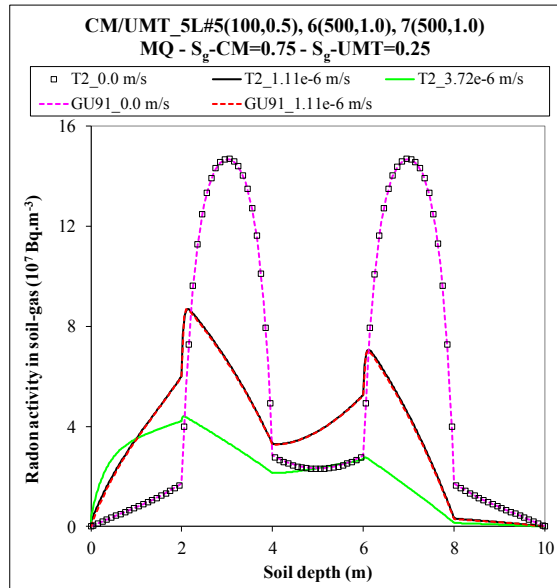


Figure 9. Numerical (T2) and analytical (GU91) profiles of radon activity in the soil-gas (CM/UMT_5L#5-7).

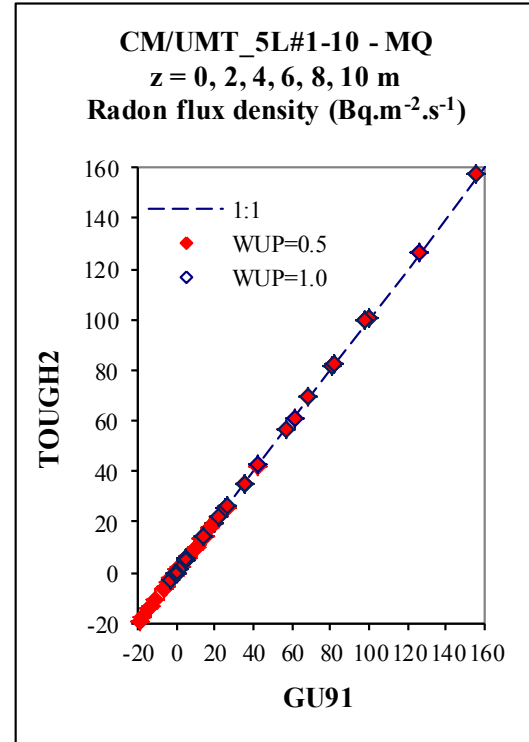


Figure 10. Numerical (TOUGH2) and analytical (GU91) radon flux densities at different depths (CM/UMT_5L#1-10).

CONCLUSION

The newly developed EOS7Rn module can now simulate radon generation by emanation, and its transport within the subsurface by accounting for all its physical properties and their eventual dependency on hydrodynamic and thermodynamic soil properties. The EOS7Rn module can be applied to groundwater and geothermal reservoir processes as well. The next task will be the *in situ* validation of EOS7Rn for radon exhalation to indoor and outdoor environments.

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